

tetrakis(perfluoroctyl) borate anion, tetrakis(perfluoro-3-methylbutyl) borate anion, tetrakis(perfluoro-5-methylbutyl) borate anion, tetrakis(heptafluoropropyl) borate anion, tetrakis(3,5-dichlorophenyl) borate anion, tetrakis(4-chlorophenyl) borate anion, tetrakis(benzyl chloride) borate anion, tetrakis(chlorobenzyl) borate anion, tetrakis[2-(perfluorobutyl)ethyl] borate anion, tetrakis[2-(perfluorohexyl)ethyl] borate anion, tetrakis[2-(perfluoroctyl)ethyl] borate anion, tetrakis[2-(perfluoro-7-methylhexyl)ethyl] borate anion, tetrakis[2-(perfluoro-5-methylhexyl)ethyl] borate anion, tetrakis[2,2,3,3-tetrafluoropropyl] borate anion, tetrakis(1H,1H,5H-octafluoropentyl) borate anion, tetrakis(1H-perfluorohexyl) borate anion, tetrakis(1,1-difluoroethyl) borate anion, tetrakis[3,5-bis(trifluoromethyl)benzyl] borate anion, tetrakis[4-(trifluoromethyl)benzyl] borate anion, tetrakis(3,5,-difluorobenzyl) borate anion, tetrakis(4-fluorobenzyl) borate anion, tetrakis(4-ethoxyphenyl) borate anion, tetrakis(4-methoxyphenyl) borate anion, tetrakis(4,5-dimethoxyphenyl) borate anion, tetrakis(4-butylphenyl) borate anion, tetrakis(t-butylphenyl) borate anion, tetrakis(phenyl) borate anion, tetrakis(biphenyl) borate anion, tetrakis(terphenyl) borate anion, tetrakis(mesityl) borate anion, tetrakis(pentamethylphenyl) borate anion, tetrakis(3,5-dimethylphenyl) borate anion, tetrakis(cyclopropyl) borate anion, tetrakis(cyclobutyl) borate anion, tetrakis(cyclohexyl) borate anion, tetrakis(cyclopentyl) borate anion, tetrakis(cyclooctyl) borate anion and tetrakis(phenoxybutyl) borate anion.

IN THE ABSTRACT:

After the claims, please insert a page containing the Abstract Of The Disclosure, which is attached hereto as a separately typed page.

REMARKS

The specification has been amended to place the application in conformance with standard United States patent practice.

Examination and allowance of pending claims 16-23 are respectfully
requested.

Respectfully submitted,

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MARKED-UP AMENDED SPECIFICATION PARAGRAPHS

Page 1, first section heading

[Technical Field] 1. Field of the Invention

Page 1, second section heading

[Background of the Invention] 2. Description of the Related Art

Page 1, third complete paragraph

High molecular weight products produced by hardening (or polymerizing) the above-mentioned compounds have various uses, especially epoxy resin and silicone resin which have been widely used as adhesives, sealing, and paints in various fields such as automobile industry, housing/building material industry, civil engineering and construction industries, aircraft industry, and electric and electronic industries.

Page 1, fifth complete paragraph

It has been well known that the polymerization is generally carried out at room temperature or under heating [condition] conditions. Recently, [the] searches [on] for a photopolymerization catalyst or initiator have been conducted, because it may be preferable to carry out the polymerization under irradiation [condition dependently] conditions depending upon use. However, [any] no remarkable result has [not] been obtained.

Page 2, fourth complete paragraph

As [other] another example, the photopolymerization initiator disclosed in Japanese Patent Publication No. 62692/94 of the same applicant as this application can be cited. This [potopolymerization] photopolymerization initiator comprises a composite obtainable from a reaction between (a) charge transfer complex consisting of biscyclopentadienyl iron derivative and quinoid and (b) at least

one salt selected from a group consisting of tetrafluoroborates, hexafluorophosphates and hexafluoroantimonates.

Page 2, fifth complete paragraph:

On the contrary, regarding [to photopolymerization] the photopolymerization of organosiloxane, it is very hard to find [out the] an example. An example can be found only in Kevin D. Belfield et al., "Photoinitiated cationic ring-opening polymerization of a cyclosiloxane", Polymer Bulletin 38, pp. 165-168 (1997). [Kevin] Belfield et al. disclosed that when hexamethylene cyclotrisiloxane is exposed with U.V. light in the presence of sulfonium or iodonium salt or iminosulfonate derivative, it is subjected to a photo-initiating cationic ring-opening polymerization in solution or in the absence of solvent.

Paragraph bridging pages 2 and 3

Regarding [to] the thermal hardening (polymerization) reaction which is carried out at room temperature or under heating [condition] conditions, as example of catalyst for polymerization of epoxy compound, tertiary amine (such as benzylmethyl amine, [2,4,6-trisdimethylamine] 2,4,6-trisdimethylamine methylphenol, etc.), imidazole (such as 2-methylimidazole, 2-ethyl-4-methylimidazole, 2-heptadecylimidazole etc.), Lewis acid (such as BF_3 monoethylamine, BF_3 piperazine, etc.) have been known.

Page 3, first complete paragraph

The thermal hardening reaction of organosiloxane compound is [almost] classified into addition type and condensation type: in a reaction of the former type, peroxide or platinum compound is used as catalyst and in a reaction of the latter type, metal salt of carboxylic acid is used.

Page 3, second complete paragraph

As mentioned above, epoxy resins and silicone resins have been widely used. However, on producing the epoxy resins or silicone resins by hardening (or polymerizing) each monomer, the catalyst or initiator used in the thermal polymerization or photopolymerization of epoxy compound is completely different from that used in the thermal polymerization or photopolymerization of organosiloxane compound, and additionally, [the] an initiator for photopolymerization of organosiloxane compound [has not] is almost [been known] unknown.

Page 3, section heading

[Disclosure of the Invention]

Summary of the Invention

Page 4, section heading

[Best Mode for Carrying out the Invention] Description of the Preferred Embodiments

Page 4, third complete paragraph which begins with "The crystalline"

The crystalline ion-association substance of the present invention comprises, as [seeing] is seen from the general formula (I), a metallocene derivative cation and a tetradeятate borate complex anion wherein the four ligands are the same to each other.

Paragraph bridging pages 4 and 5

In the general formula (I), the electron donative or electron attractive substituent is alkyl group, cycloalkyl group, aryl group, alkoxy group, silyl group, dialkyl group, acyl group, cycloalkenyl group, amino group, carboxyl group, [organobolyl] organoboranyl group, phosphino group, aldehyde group, hydroxyl group and the like. More particularly, the alkyl group is selected from among lower alkyl groups such as methyl group, ethyl group, propyl group and butyl group, or

pentyl group or amyl group; and the cycloalkyl group is selected from among cyclobutyl group, cyclopentyl group, cycloheptyl group, and cyclohexyl group. The aryl group is selected from among phenyl group, naphtyl group, etc.

Pages 8, first complete paragraph

The crystalline ion-association substance of the present invention contains a bulky ligand in the [cyclopentadienyl] cyclopentadienyl moiety of the metallocene derivative cation. Therefore, [the] a transition metal of the center nucleus [under] having a high oxidation state may maintain thermal stability, and the crystallinity between the metallocene derivative cation and the anion complex to be associated is synergistically increased.

Page 11, first complete paragraph

The crystalline ion-association substance of the present invention may have such a structure that one of four identical ligands of the borate complex anion is closed to the transition metal center of the metallocene derivative cation and is sandwiched between the two cyclopentadiene ligands of the metallocene derivative cation; the metallocene derivative cation per se having such a structure that the two cyclopentadiene ligands with various structure are positioned in the form of [dihederal] a dihedral structure with respect to the transition metal center.

Paragraph bridging pages 11 and 12

In photopolymerization of a cationically polymerizable organic substance, the polymerization initiator of the present invention is used in the amount of from 0.1 to 10 weight parts, preferably 0.5 to 4 weight parts with respect to 100 weight parts of the cationically polymerizable organic substance; when the cationically polymerizable organic substance absorbs usually 2000 to 9000 mJ/cm² of energy by irradiation with U.V. light having the wavelength of 200-700nm, basically

200-400nm, a high molecular weight polymer is produced. However, [the] an irradiation operation with more energy may be required depending upon the type of polymerizable substance. The irradiation can be carried out at any condition, that is, at ordinary temperature, under cooling condition, or under heating condition; and at atmospheric pressure or vacuum, or in the presence of inert gas.

Page 12, third complete paragraph

When compatibility between the cationically polymerizable organic substance and the polymerization initiator is poor, a suitable solvent such as hydrocarbon solvents, halogenated hydrocarbon solvents, alcoholic solvents, phenolic solvents, ether/acetal solvents, ketone solvents, ester solvents, nitrogen-containing compound solvents may be used. The concrete examples of the solvent are dichloromethane, chloroform, methanol, tetrahydrofuran, acetone, [metylethyl] methylethyl ketone, [acetonitorile] acetonitrile, etc. The solvent is used in 5-3 times amount with respect to 100 weight parts of the cationically polymerizable organic substance, that is, 500-300 weight parts.

Page 17, subparagraph III) (b)

(b) the hardened product has the substantially unchanged characteristics of the starting material itself; for example, when it is mixed with the [cationically] cationically polymerizable organic substance such as epoxy compound, the obtainable polymer product (hardened product) has the unchanged inherent characteristics of epoxy compound, and additionally, it is possible to produce composite product (hardened product) consisting of at least two cationically [polymerizable] polymerizable organic substances which are of [the] a different type from each other.

Page 18, second paragraph

In preparation of the crystalline ion-association substance of the present invention, commercially available products (for example, products sold by Tokyo Chemicals Co. or [Aldolich] Aldrich Co.) can be used as metallocene derivative. Other metallocene derivatives can be prepared by the well-known methods for preparation disclosed in the following references:

Page 22, first complete paragraph

By using [the] a similar apparatus and procedure to those [ised] used in [the] Example 4, 0.46g of (dihydroxyboryl) ferrocenium/tetrakis(3,5-difluorophenyl) borate was obtained (yield 51%), starting from 0.3g of commercially available ferroceneboronic acid (CAS 12152-94-2), 0.021g of iron(III) chloride, 30ml of acetonitrile, 10ml of pure water and 0.635g of tetrakis (3,5-difluorophenyl) borate sodium salt.

Paragraph bridging pages 26 and 27

On the other hand, in case of using an organosiloxane compound having volatility and subliming property, a test sample was prepared as [follpws] follows:

In [the] a similar bottle to that described above, 10 weight parts of dichloroemethane is added with respect to 100 weight parts of the organosiloxane compound, and then the predetermined amount of polymerization initiator is added thereto. The resulting liquid mixture is stirred to disperse the initiator. Thereafter, the liquid mixture enclosed in the bottle is irradiated repeatedly with U.V. light (6000mJ/cm²) four times from [outer] outside of the bottle. The obtained test sample (rubber-like hardened product produced in the bottle) is taken out and transferred to a glass cylinder for

Soxhlet extraction test (glass filter with filter plate No. GP-100) to measure gel fraction.

Page 32, second complete paragraph

To 100 weight parts of decamethylcyclopentasiloxane (D5) or [dodecamethylcyclohexasiloxane] dodecamethylcyclohexasiloxane (D6), 2 weight parts of each polymerization initiator which comprises the crystalline ion-association substance prepared in the Example 8 or 10, 4 weight parts of benzophenone (sensitizer) and 30 weight parts of dichloromethane were added. The obtained mixture was stirred and mixed to prepare a solution in which the organosiloxane compound, the sensitizer and the polymerization initiator were homogeneously dissolved in the solvent. The obtained solution was fed to the center recess of the hole slide glass of one hole type without overflowing the solution from the recess. The hole slide glass was put into a hot air circulating oven and maintained at 60°C for 10 min. The test film was cooled to room temperature and irradiated with U.V. light (600 mJ/cm²). The gel fraction of the hardened films was reported in the following Table 4.

Page 33, second complete paragraph

To 100 weight parts of 1,3-bis(glycidoxypropyl) tetramethyldisiloxane (silicone-modified epoxy monomer) (Gelest Inc.; SIH1115.0; CAS 126-80-7), 1 weight part of each polymerization initiator which comprises the crystalline ion-association substance prepared in the Example 2, 3, 8, 9, 10, 11, 12 or 13, 2 weight parts of benzophenone (sensitizer) and 50 weight parts of dichloromethane were added. The obtained mixture was stirred and mixed to prepare a solution in which the silicone-modified epoxy compound, the sensitizer and the polymerization initiator were homogeneously dissolved in the solvent. The obtained solution was fed to the

center recess of the hole slide glass of one hole type without overflowing the solution from the recess. The hole slide glass was put into a hot air circulating oven and maintained at 60°C for 10 min. and then at 80°C for 3 [mine] min. Each test film was irradiated with U.V. light (8000mJ/cm²).

Page 40, first complete paragraph

These examples show the fact that the polymerization initiator which comprises the crystalline ion-association substance prepared in the above-mentioned Example 3, 9, 11 or 12 has an ability to [proceed] produce a thermal polymerization of a mixture consisting of cyclic siloxane and epoxy compound.

Page 42, first complete paragraph beginning with "The polymerization"

The polymerization initiator of the present invention [have such a] has the unique property that it can initiate both photopolymerization and thermal polymerization when used in polymerizing cationically polymerizable organic substance, though the polymerization initiator comprises an identical crystalline ion-association substance.

Page 42, second complete paragraph beginning with "In particular"

In particular, since the photopolymerization initiator for organosiloxane has [not almost] been little developed, the polymerization initiator of the present invention is valuable.

Paragraph bridging pages 42 and 43

For the above-mentioned unique property of the polymerization initiator of the present invention, it is possible to obtain a merit in practical use; that is, after the U.V. light irradiation, it is possible to carry out the hardening in a shade zone which is not exposed to U.V. light or a deep zone to which U.V. light [dose] does not reach by means of heating (photopolymerization plus thermal

polymerization). This merit is very important, because, in accordance with conventional method, when the hardening is carried out by using both means of photopolymerization and thermal polymerization in accordance with conventional method, it is necessary to modify a monomer itself; for example, to prepare a modified organosiloxane having epoxy group.

Page 43, last complete paragraph

Therefor, it is possible to [spread] extend the field and method for use of cationically polymerizable organic substances by using the polymerization initiator of the present invention.